The effect of magnetic (phase) inhomogeneities upon the low temperature conductivity of antiferromagnetic $La_2CuO_{4+\delta}$ single crystal

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It is found that a strong inhomogeneous distribution of oxygen in single crystal $La_2CuO_{4+\delta}$ can lead to the formation of isolated superconducting inclusions, the average hole concentration being no more than 0.0024 per Cu atom. The behavior of the conductivity in the magnetic field below $T = 20$ K is consistent with possible existence of a insulating low-temperature magnetic phase (spin density waves), which is typical of cuprates with excess oxygen.

Properties of strongly correlated electronic systems are greatly affected by local fluctuations and nonuniform distribution of the charge and spin density. In this respect, the behavior of high-temperature superconductors (HTSCs) belonging to the family of lanthanum cuprates can be considered as unique. Such behavior is often due to the phase separation (typical of these systems) into regions with different charge-carrier concentrations and the corresponding local disturbances of structural and magnetic order.

In this study, we revealed a possible effect of spatially nonuniform distribution of excess oxygen on the low-temperature behavior of the conductivity of a $La_2CuO_{4+\delta}$ single crystal in the antiferromagnetic state. The initial $La_2CuO_{4+\delta}$ single crystal had the Néel temperature $T_N \approx 266$ K and a hole concentration of about 0.0025 per copper atom (according to the estimations based on the data of Ref. [1]). X-ray diffraction analysis revealed the presence of twins, which inevitably arise upon cooling of a crystal below the point of structural phase transition (530 K) from the tetragonal phase to the orthorhombic phase [2]. At low voltages, the current-voltage (I-V) characteristics of this sample have a relatively narrow ohmic portion, in which the Mott's law of variable-range hopping for three-dimensional systems is satisfied in the temperature range 20–250 K [3]:

$$R \propto \exp\left(\frac{T_0}{T}\right)^{1/4}, \tag{1}$$

The localization length L_c can be found from the T_0-value [3]. Our estimate for the CuO_2 plane is $L_c \approx 0.4$ nm. At $T < 20$ K, the dependences $R(T)$ deviated from the Mott's law towards lower resistance.

The initial sample was kept at room temperature in air for 3 years. One might expect the aging of such kind lead to a significant redistribution of oxygen in the crystal due to the high mobility of excess oxygen and the tendency of this system to phase separation. Since holes in $La_2CuO_{4+\delta}$ are basically of oxygen origin [4], this process should also lead to nonuniform distribution of the charge carrier density and local break of the antiferromagnetic order within and near the boundaries of hole-enriched regions (arising due to the phase separation, inherent in doped cuprates [4]). The effect of such inhomogeneities on the transport properties of the $La_2CuO_{4+\delta}$ sample is considered below.

As a result of the 3-year exposure of the sample, its Néel temperature has increased to 269 K and the hole concentration lowered to 0.0024. The minor difference in T_N between the starting and aged samples points to a small decrease in the concentrations of oxygen and charge carriers. This leads us to assume that the observed changes in the behavior of the transport properties are mainly due to the redistribution of oxygen and charge carriers over the crystal volume rather than to a change in the average hole concentration.

The resistance along CuO_2 planes was measured with a constant current through the sample. The current was varied from 0.03 to 2 μA. The temperature dependences $R(T)$ in the range 5-100 K at several currents are shown in Figs. 1a and 1b. As the temperature lowers to 45 K, the curves deviate from the Mott's law (Fig.1a) towards lower resistance. At $T < 25$ K and $J \leq 0.2$ μA there is a change to a simple activation dependence $R \propto \exp(\Delta/kT)$, where $\Delta = 32.4$ K (Fig.1b). Above 45 K, the dependence $R(T)$ at a current 0.2 μA is described by Eq. (1) with $T_0 \approx 2.3 \times 10^6$ K, which corresponds to the localization length $L_c \approx 0.262$ nm. The obtained value of L_c is much smaller than the orthorhombic lattice parameters in a CuO_2 plane ($a = b = 0.54$ nm). This points out to a much stronger localization and less uniform distribution of charge carriers in comparison with the initial state.

The behavior of $R_{ab}(T)$ in the temperature range $T < 50$ K is nonohmic; it demonstrates a strong dependence of the resistance on current. At some critical current $J > 0.2$ μA, the resistance sharply decreases (Fig. 1a). The changes in $R(T, J)$ correlate with the behavior of the I-V characteristics in the temperature range 5-80 K. Typical behavior of the I-V characteristics and the corresponding dependences of the resistance on current is shown in Figs. 2a and 2b. The dependences $U(J)$ and $R(J)$ are essentially nonlinear and have a region with current-controlled negative differential resistance. The characteristic currents J_1 and J_2 are marked

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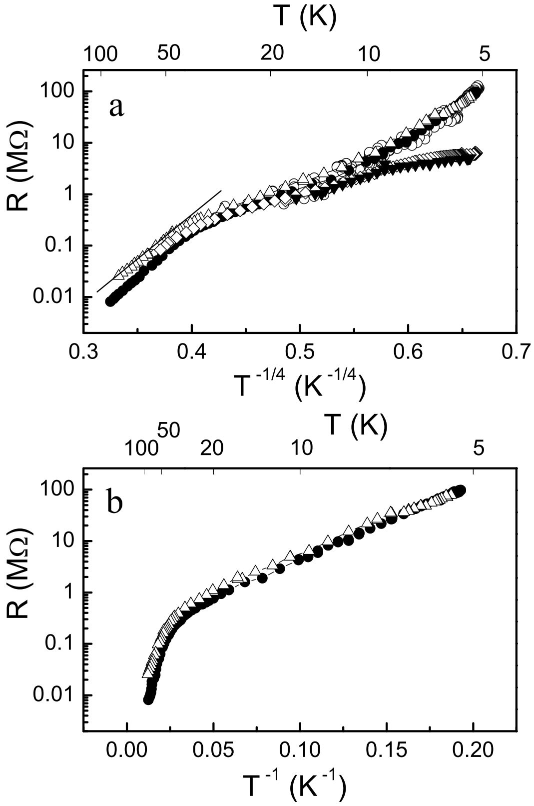

FIG. 1: (a) Temperature dependence of the resistance lg R *vs.* $T^{-1/4}$ of a $La_2CuO_{4+\delta}$ single crystal ($T_N \approx 269$ K) at different measuring currents: (○) 0.03, (•) 0.06, (△) 0.2, (◇) 0.5, and (▼) 2μA; (b) the dependence lg R *vs.* T^{-1} for currents of (•) 0.06 and (△) 0.2 μA.

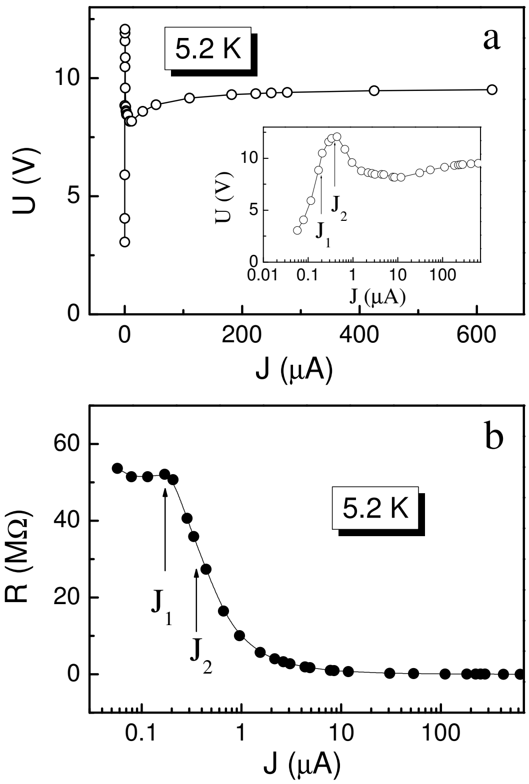

FIG. 2: (a) Current-voltage characteristic of the $La_2CuO_{4+\delta}$ single crystal at $T = 5.2$ K. Inset: the same dependence in the semilogarithmic coordinates; (b) the corresponding dependence of the resistance on current.

with arrows. The current J_1 corresponds to the inflection point in the ascending portion of the dependence $U(J)$ (Fig. 2a, inset) and the crossover point in the curve $R(J)$ (Fig. 2b). The current J_2 corresponds to the maximum in the dependence $U(J)$ and the inflection point in the lower branch of the curve $R(J)$. These features of the I-V characteristics are observed at all temperatures in the range 5-80 K. Comparison of the I-V characteristics with the temperature dependences of the resistance shows that the character of the dependence $R(T)$ changes at $J = J_1$. A simple exponential dependence is observed at currents $J \leq J_1$ ($J_1 = 0.2$ μA). Deviation from the law $R \propto \exp(1/T)$ (see Fig. 2) occurs at currents $J > J_2$ ($J_2 = 0.44$ μA).

The behavior of the magnetoresistance in the $La_2CuO_{4+\delta}$ single crystal studied is also dependent on the transport current in the range 5-80 K. The magnetoresistance is positive for low currents in the range $T \leq 17$ K. Positive magnetoresistance is observed at temperatures and currents for which the law $R \propto \exp(\Delta/kT)$ is obeyed. The magnetoresistance becomes negative with an increase in the temperature or current (Fig. 3). Above 17 K, only negative magnetoresistance is observed, whose magnitude decreases with an increase in current.

It is known that, in the regime of hopping conductivity in sufficiently low electric fields ($E \ll kT/er_h\gamma$), the resistance is field-independent (here, r_h is the mean hopping distance and γ is a numerical factor of the order of unity). According to the estimation performed, this is true to a great extent for the sample under study; therefore, the observed nonlinear behavior of the I-V characteristic (Fig. 2) cannot be directly attributed the effect of electric field on the hopping conductivity of a homogeneous system. At the same time, the weak dependence of the resistance on current at $J \geq 1$ μA suggests the absence of Joule heating.

The dependence $R \propto \exp(\Delta/kT)$ observed in the low temperature range suggests that the oxygen disordering produces a gap ($\Delta = 32.4$ K) in the spectrum of quasiparticle excitations involved in the charge transfer. It is interesting to clarify the nature of this gap.

The revealed changes in the behavior of hopping conductivity below 25 K correspond well to the known phenomena in inhomogeneous systems (mixtures of insulating and superconducting phases). Cooling of such a sys-

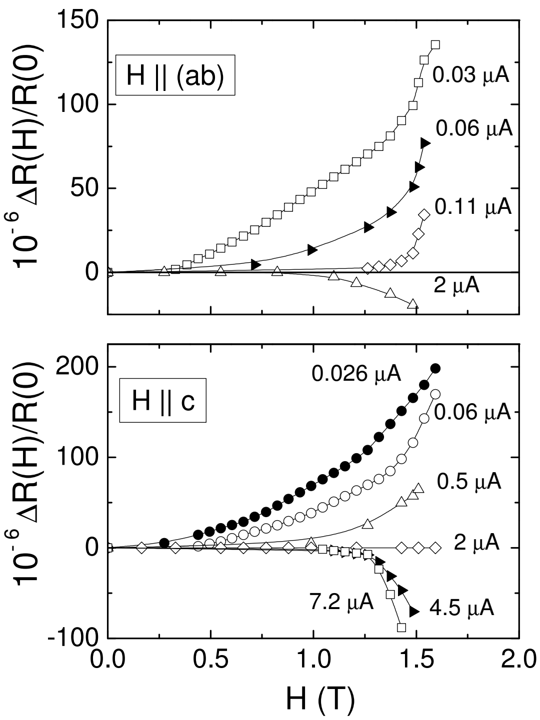

FIG. 3: Magnetoresistance curves taken at $T = 5$ K for different transport currents $J \parallel (ab)$ for two the magnetic field directions: $\mathbf{H} \parallel (ab)$ and $\mathbf{H} \parallel \mathbf{c}$.

tem below T_c causes a transition from the variable-range hopping to the dependence $R \propto \exp(\Delta/kT)$, where Δ is a superconducting gap, playing the same role as the band gap in insulators [5, 6]. In our case, the formation of a superconducting phase can be related to the phase separation in lanthanum cuprates [4, 7], which yields two phases with $\delta = 0$ and $\delta > 0$. Previously, the transition to a simple exponential dependence in $La_2CuO_{4+\delta}$, caused by nonuniform oxygen distribution, was observed at $T < 20$ K for a far less resistive sample of $La_2CuO_{4+\delta}$ with $L_c \approx 1$ nm [8] and a hole concentration of 0.0044. The sample studied has a much lower average hole concentration and a shorter localization length (0.26 nm). At the same time, it is characterized by a larger degree of inhomogeneity due to the more nonuniform oxygen distribution as a result of the long-term exposure (aging). In our opinion, it is the phase separation that determines the observed features of low-temperature behavior of the resistance of the sample studied. With a decrease in temperature, the oxygen-enriched isolated regions turn to the superconducting state. As a result of the decrease in the density of single-particle excitations and weak coupling between isolated superconducting inclusions at sufficiently low temperatures, a transition from dependence (1) to a stronger simple exponential dependence occurs. An increase in current leads to depairing of carriers and, possibly, their heating under conditions of strong inhomogeneity. Either of the mechanisms should lead to a decrease in the resistance, which is observed at currents $J > 0.2$ μA.

The suppression of local superconductivity by the magnetic field is also expected to break the pairs of charge carriers and reduce resistance [5]. We however observed positive magnetoresistance in the low temperature region at quite small currents, which is rather unexpected result. This behavior of magnetoresistance can be attributed to the new low-temperature magnetic phase recently detected in $La_2CuO_{4+\delta}$. The investigation of phase separation in lanthanum cuprates with excess oxygen shows that the dielectric phase in the mixed system below $T = 40$ K is a new magnetic state, namely, a spin-density wave [9] rather than the Néel phase. The relative volume of this phase increases with a decreasing in content of excess oxygen (with correspondin decrease in T_c). Therefore, in lightly doped $La_2CuO_{4+\delta}$ compound, this phase is rather abundant and the content of the superconducting phase is much smaller. The magnetic field stabilizes the magnetic phase by reducing the volume fraction of the superconducting phase and thus imitates a change in the doping level [9]. The effect is not yet clear completely but it is obvious that in this case the magnetic field should induce positive magnetoresistance.

In conclusion, the results of this investigation indicate that, under conditions of strong inhomogeneity of a sample at temperatures $T < 25$ K, phase separation even in $La_2CuO_{4+\delta}$ with a low content of excess oxygen (hole concentration not higher than 0.0024 per copper atom) may lead to the formation of isolated regions (clusters) of a superconducting phase. The magnetoresistance behavior of the sample in this temperature interval can be attributed to the presence of the insulating magnetic phase (spin density wave), as assumed in [9].

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